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Magnetic moments in a gadolinium iron garnet studied by soft-X-ray magnetic circular dichroism

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The magnetic moments of Gd and Fe in gadolinium iron garnet ($\text{Gd}_3\text{Fe}_5\text{O}_{12}$) were probed at 77 and 300 K by soft-X-ray magnetic circular dichroism (SXMCD) measurements at the $\text{GdM}_{4,5}$ and at the $\text{FeL}_{2,3}$ absorption edges. The SXMCD signal at each edge allows one to independently determine the magnetic ordering for each specific ion, and the temperature dependence confirms that the reversal of the macroscopic magnetization is due to the reversal of each local magnetic moment of the Fe and the Gd atoms when the compensation point ($T_{\text{comp}} = 288$ K) is crossed. This work therefore demonstrates that SXMCD can probe element and site specific magnetic properties of multi-component systems.

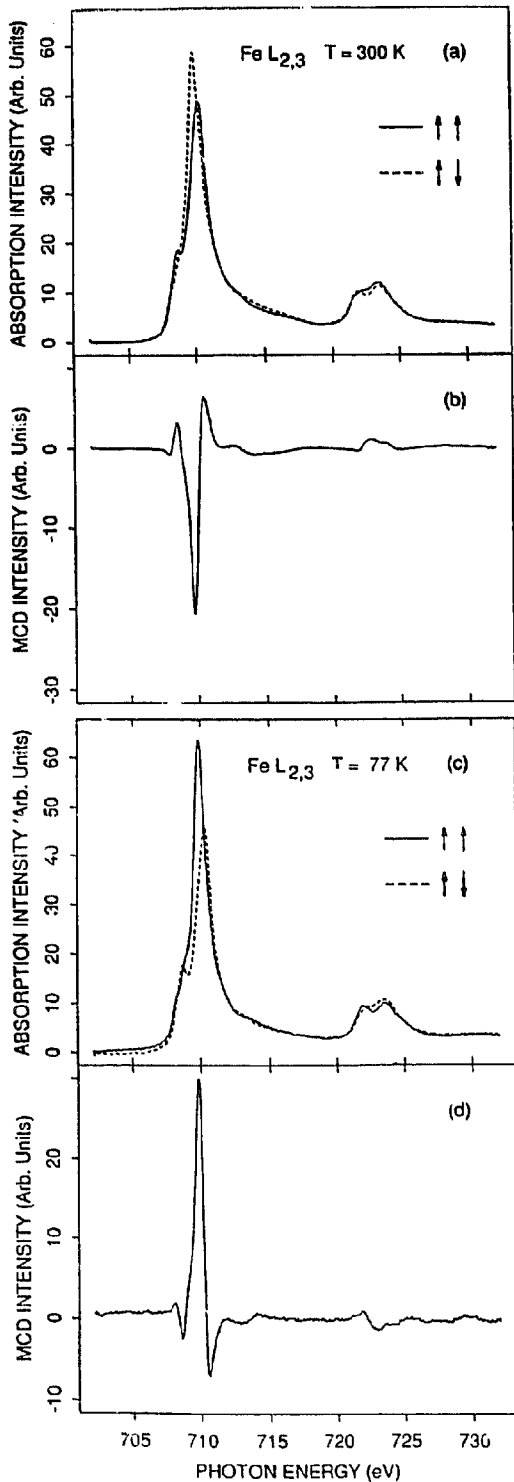
Circularly polarized synchrotron radiation, so far accessible in the ultraviolet [1] and in the hard-X-ray region [2], has recently become available also in the soft-X-ray region [3,4]. This opens up new opportunities for magnetic circular dichroism (MCD) studies with the dipole permitted $2p \rightarrow 3d$ and $3d \rightarrow 4f$ excitations in 3d transition metal and 4f rare earth magnetic systems. The soft-X-ray magnetic circular dichroism (SXMCD) observed in near edge core absorption processes is related to the magnetic moment of the photoexcited atom when the core electron is promoted into final states that are responsible for the ferro- or ferri-magnetic properties of the system. Simple exchange-split-valence-band [5] and atomic [6] models have been proposed previously to interpret core-excited MCD spectra, and provide a basic tool for determining the element specific magnetic ordering.

In this work we utilized SXMCD to investigate the magnetic properties of gadolinium iron garnet, $\text{Gd}_3\text{Fe}_5\text{O}_{12}$. The $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ has the following interesting magnetic structure [7]: there are

two sublattices of ferric ions, antiferromagnetically coupled by superexchange through oxygen atoms, which respectively contain 3 Fe^{3+} per formula unit on tetrahedral sites and 2 Fe^{3+} per formula unit on octahedral sites. The Gd^{3+} ions are also antiferromagnetically coupled through superexchange to the net moment of the iron ions, but this coupling is much weaker than that among iron ions. As a consequence, at room temperature the Fe lattices dictate the ferrimagnetic properties of the material and the Gd ions are essentially disordered. At lower temperatures, however, the Gd lattice also becomes ordered. Since the Gd magnetic moment (approximately $21\mu_B$ per formula unit) is much larger than the net magnetic moment of the Fe lattices (approximately $5\mu_B$ per formula unit), there exists a certain temperature, the compensation point (288 K) [8], at which the macroscopic magnetization flips direction.

The aim of this work is to investigate if the accepted local magnetic moment picture of this material is directly reflected by the SXMCD signal at the Fe and Gd edges and therefore to establish if the SXMCD is indeed directly related to the magnetic moment of the ion.

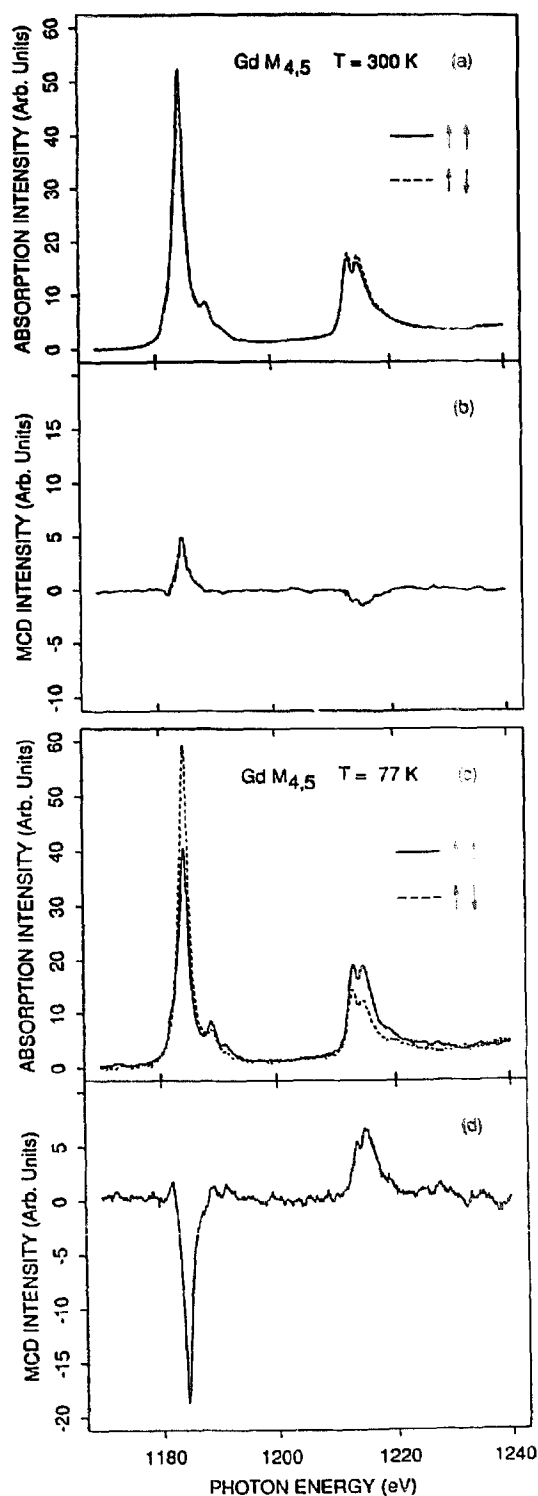
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The experiments were carried out using the AT & T Bell Laboratories Dragon high resolution soft-X-ray beamline at the National Synchrotron Light Source [9]. The optical configuration of this beamline which allows the selection of circularly polarized synchrotron radiation above and below the orbital plane of the storage ring has been described elsewhere [3]. For the measurements discussed in the following, the monochromator resolution was set to 0.6 and 0.3 eV at the Gd $M_{4,5}$ and Fe $L_{2,3}$ edges respectively. The degree of circular polarization was calculated to be $\sim 80\%$ and $\sim 90\%$ operating at an angle away from the orbit plane $\psi_{\text{off}} = 0.5$ mrad at the Gd $M_{4,5}$ edges and $\psi_{\text{off}} = 0.75$ mrad at the Fe $L_{2,3}$ edges, respectively. The vertical angular acceptance of the monochromator was set to $\Delta\psi = 0.25$ mrad in both cases. The photoabsorption spectra were recorded at normal incidence on a $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ single crystal by monitoring the sample photocurrent. The magnetic field at the sample surface was ~ 4 kG. The parallel or antiparallel orientation between the external magnetic field and the circular polarization was accomplished by flipping an external permanent magnet.

The Fe $L_{2,3}$ photoabsorption spectra taken for two different magnetization directions at 300 K are displayed in fig. 1a and the corresponding SXMCD data are shown in fig. 1b. Previous studies of the SXMCD effect in a Fe single crystal [4] have shown a reversed sign of the SXMCD signal between the L_3 and the L_2 edges and a negative dichroism at the L_3 edge; the same behavior is predicted by a recent calculation [10] of the magnetic circular dichroism in the 2p absorption spectra of a $3d^5$ transition metal ion. This finding gives us a key to the understanding of the SXMCD signal observed for the garnet: what we see is the

Fig. 1. Fe $L_{2,3}$ absorption spectra measured with circularly polarized soft-X-rays in a magnetically oriented $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ single crystal for two different orientations of the external magnetic field at 300 K (a) and 77 K (c). The magnetic circular dichroism spectra, (b) and (d), are the difference between the solid and the dashed curves shown in (a) and (c), respectively.



superposition of the independent contributions from the Fe^{3+} ions in the two sublattices. The tetrahedral lattice is the majority one with the magnetic moment aligned along the external field. It contributes with a negative signal at the L_3 edge and a positive one at the L_2 edge. The octahedral sites, being antiferromagnetically coupled, can be associated with the positive signal at the L_3 edge. The understanding of the detailed lineshape and intensity requires more sophisticated calculations in which the chemical shifts and the crystal fields effect in the respective octahedral and tetrahedral symmetry are included.

Figs. 1c and d show the $\text{Fe } L_{2,3}$ photoabsorption and SXMCD spectra taken at 77 K. When the temperature is lowered to 77 K, the magnetization becomes dominated by the Gd magnetic moment and consequently the orientation of the two Fe sublattices is reversed with respect to the 300 K case. This is indeed confirmed by the Fe SXMCD signal. The absolute SXMCD intensity changes by a factor of 1.5 with temperature, as one expects from the earlier calculations by Pauthenet [11] on the temperature dependence of the magnetic moments of the various ions in a gadolinium iron garnet.

A more pronounced temperature dependence is observed in the Gd $M_{4,5}$ edges. Figs. 2a and b show the absorption and MCD spectra taken at 300 K, and figs. 2c and d those for $T = 77$ K. At 300 K the dichroism is positive at the M_5 edge and negative at the M_4 edge. Similar to the Fe case, the sign of the magnetic circular dichroism can be related to the orientation of the magnetic moment of Gd by a simple atomic model for the states [4,6]. One expects a negative MCD at the M_5 and a positive MCD at the M_4 edge for $3d \rightarrow 4f$ transitions if the atom's magnetic mo-

Fig. 2. Gd $M_{4,5}$ absorption spectra measured with circularly polarized soft-X-rays in a magnetically oriented $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ single crystal for two different orientations of the external magnetic field at 300 K (a) and 77 K (c). The magnetic circular dichroism spectra, (b) and (d), are the difference between the solid and the dashed curves shown in (a) and (c), respectively.

ment is aligned parallel to the external magnetic field. The observation of a positive MCD signal at the M_5 edge therefore confirms a residual anti-ferromagnetic coupling between the Gd^{3+} ions and the net Fe lattices' magnetic moment. As shown in figs. 2b and d, at low temperatures the dichroism changes sign, in agreement with the reversal of the local magnetic orientation when the compensation point is crossed, and becomes much more intense (factor of ~ 4) than at 300 K. Calculations of the $M_{4,5}$ SXMCD of an isolated Gd^{3+} ion [6] reproduce very well the general features of the experimental spectra, but do not include a temperature dependence.

In conclusion, our results demonstrate that the relative magnitude and orientation of local magnetic moments of specific atoms can be determined from soft-X-ray magnetic circular dichroism measurements. The detailed interpretation of the data and their temperature dependence requires more comprehensive calculations for the photoabsorption process where both solid state and magnetic interactions are taken into account.

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